

Rapid Prototyping of Continuous Fiber Reinforced Ceramic Matrix Composites

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ABSTRACT

For ceramics to be used as structural components in high temperature applications, their fracture toughness is improved by embedding continuous ceramic fibers. Ceramic matrix composite (CMC) materials allow increasing the overall operating temperature, raising the temperature safety margins, avoiding the need for cooling, and improving the damping capacity, while reducing the weight at the same time. They also need to be reliable and available in large quantities as well.

In this paper, an innovative rapid prototyping technique to fabricate continuous fiber reinforced ceramic matrix composites is described. The process is simple, robust and will be widely applicable to a number of high temperature material systems. This technique was originally developed at the University of Delaware Center for Composite Materials (UD-CCM) for rapid fabrication of polymer matrix composites by a technique called automated tow placement or ATP. The results of mechanical properties and microstructural characterization are presented, together with examples of complex shapes and parts. It is believed that the process will be able to create complex shaped parts at an order of magnitude lower cost than current CVI and PIP processes.

INTRODUCTION

Monolithic ceramics lack the fracture toughness necessary to be considered for many high temperature applications. To be used in such applications, materials must possess low density, high elastic modulus, a low thermal-expansion coefficient, high thermal conductivity, excellent erosion and oxidation/corrosion resistance, and flaw-insensitivity. They will in many cases also be required to be joined, survive thermal cycling and multi-axial stress states, and for reusable applications the materials must maintain the above attributes after prolonged exposure

to extremely harsh chemical environments. The final and possibly most important attributes for these materials are the need to be of low cost and readily available in large quantities.

Fracture toughness of such materials could be improved by embedding continuous fibers into a ceramic matrix. This engineered material is called a ceramic matrix composite or CMC [1]. However, current state of the art methods for the fabrication of CMC materials such as chemical vapor infiltration (CVI) or polymer impregnation and pyrolysis (PIP) are expensive and time-consuming. Additionally, these processes cannot be easily adapted to rapid prototyping techniques, which would effectively reduce costs and fabrication times.

Advanced Ceramics Research, Inc. (ACR) has developed a low cost, flexible-manufacturing process for ceramic based continuous fiber reinforced composites [2]. This process, called Continuous Composite Co-extrusion (C^3) incorporates fiber tows into a ceramic matrix to fabricate ceramic-matrix/ceramic fiber composites. The C^3 process is the continuous extrusion of matrix and interface materials around a fiber tow. A flow chart illustrating the C^3 process is shown in Figure 1. A low shear strength interface material such as graphite or boron nitride is introduced between the fibers and the matrix to minimize the thermal expansion mismatch between the fibers and the matrix during the final consolidation step. Depending on the operating and processing conditions, the interface used might be multi-layered, solid, liquid or in paste form. The process is simple, robust and is widely applicable to a number of ceramic composite material systems such as Cf/SiC, Cf/ZrC, SiC/SiC etc.

The process begins by mixing the ceramic and graphite powders individually with thermoplastic binders and additives in a high shear mixer. The resulting ceramic powder/thermoplastic blend is pressed into a "green" rod. Next, the graphite powder/thermoplastic blend is pressed into a small diameter "green" rod. A hole of equal diameter to the graphite rod is drilled into the center of the rod and the graphite/thermoplastic rod is inserted. This green composite feedrod is then re-pressed and a hole is drilled in the center of the matrix/interface composite feedrod allowing the carbon fiber tow to pass through the center. Since the matrix and the interface are monolithic powders, the cost of the final composite could be lower than CVI and PIP based CMC's by at least an order of magnitude.

Recently, an innovative manufacturing technique called Advanced Tow Placement (ATP) has been developed Gillespie and co-workers [3] for thermoplastic and thermoset based polymer composite components at the University of Delaware Center for Composite Materials (UD-CCM). The ATP process is an enabling technology, developed originally for thermoset composites and more recently for in-situ non-autoclave consolidation of large-scale thermoplastic composite materials for High Speed Civil Transport (HSCT) applications. In this process, consolidation of the thermoplastic composite tows is achieved by the concurrent use of localized heat and pressure so that bonding is achieved at the newly formed interface and consolidation is achieved within the material. Figure 2 shows the schematic of the automated thermoplastic tow-placement process at UD-CCM.

Since thermoplastic binders are used in the fabrication of ACR's C^3 green CMC filaments, the ATP process originally developed for thermoplastic prepreg tows was adapted for ceramic matrix composites. The ATP process for thermoplastic prepregs was modified to lay down ceramic tows reinforced with continuous fibers in the desired configuration. This process also allows the possibility to create complicated and large parts directly from a CAD drawing without human intervention. This process is called Ceramic Composite Advanced Tow Placement or CCATP process. In this paper, initial results with the CCATP process are presented.

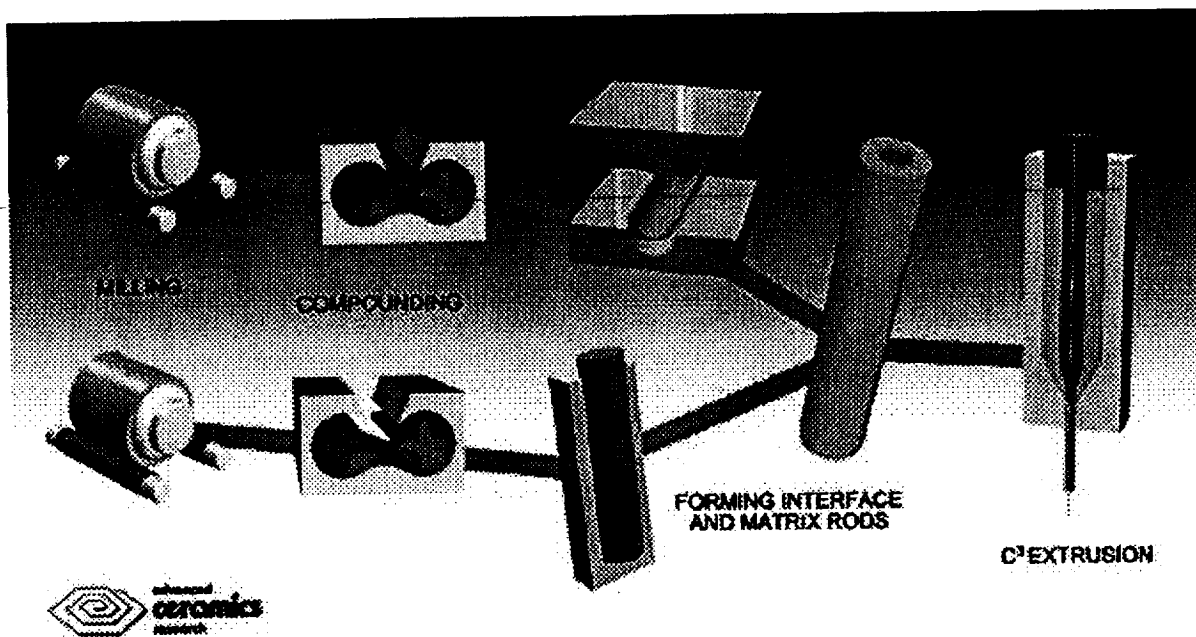


Figure 1. Flow Chart Illustrating the C³ Process.

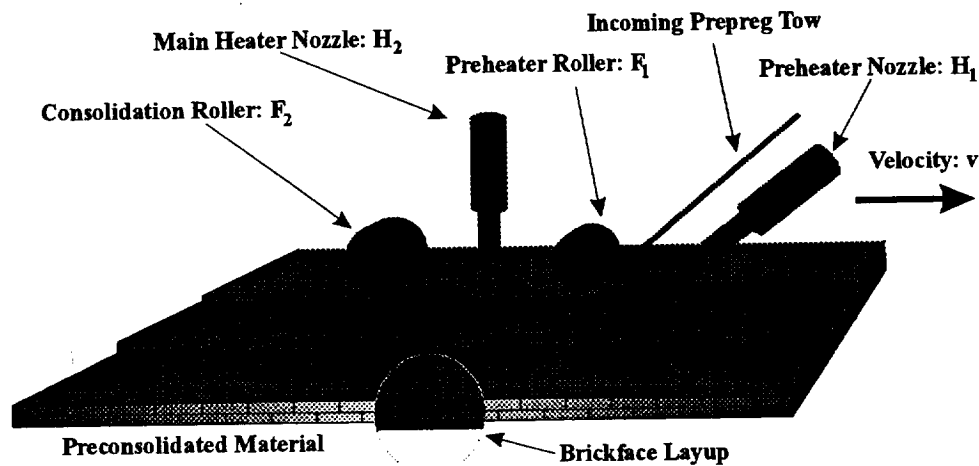


Figure 2. Schematic of the ATP process

EQUIPMENT AND EXPERIMENTAL PROCEDURES

EQUIPMENT

The automated tow placement system at the University of Delaware Center for Composite Materials was developed for rapid, low cost fabrication of fiber reinforced thermoplastic parts, and is shown in Figure 3. It employs two hot-gas nitrogen torches to heat the material and two rollers to provide the pressures required for consolidation, as shown in Figure 2. The purpose of the first torch and roller is to preheat the composite surface and incoming tow together. The material is thus "tacked" to the surface with this roller. This tacking procedure is useful in that the fed material is carefully bonded to the surface and not pulled with the main consolidation roller. This tacking approach also improves the efficiency of the cut and refeed

mechanism. The second torch (main heater) provides supplemental through thickness heating to facilitate consolidation and bonding of the tow and substrate under the consolidation roller. These rollers provide the necessary forces to achieve complete intimate contact across the tow interface, and as a boundary pressure for preventing any internal void development.

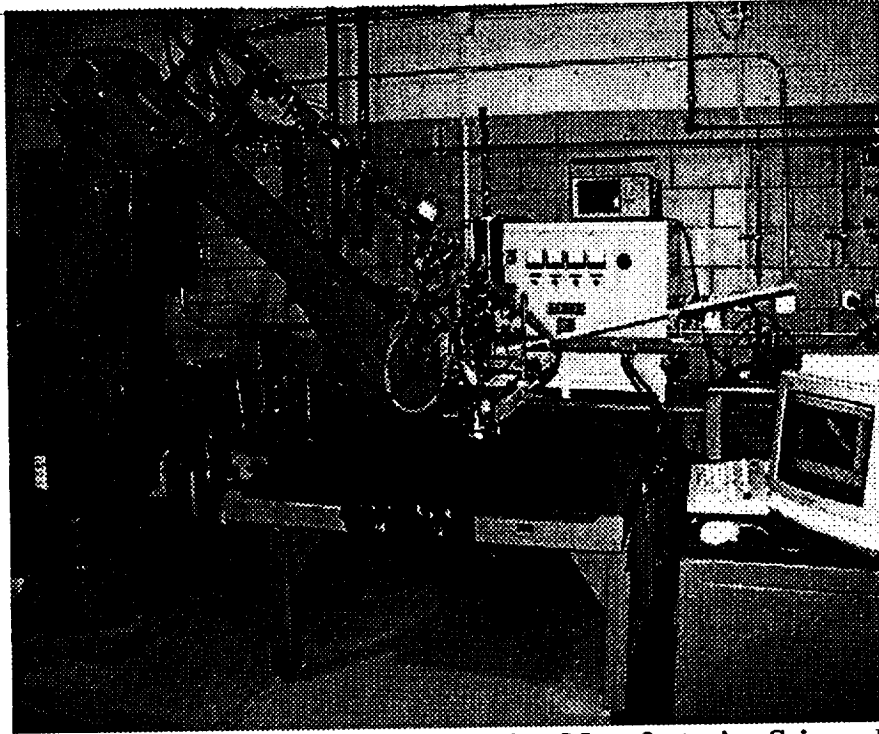


Figure 3. ATP assembly at the UD/CCM Composites Manufacturing Science Laboratory.

The forces applied to both rollers are controlled independently using a series of pneumatic actuators. The composite tows can be placed in a regular repeating pattern or with brick-face symmetry. The brick-face geometry has the advantage that more homogeneity is achieved throughout the composite structure. All the processing inputs are controlled either manually or through a PID control scheme from a LabVIEW™ interface. An ABB IRB 6400 Robotic Work cell, also shown in Figure 3, is used to control the motion of the placement head. This robot is capable of carrying a 120-kg payload at automated speeds up to 7m/s and is accurate to 0.01mm. A computer-controlled nitrogen hot gas torch control system (not shown) is used to monitor and control gas flow rate and temperatures within both torches. The composite surface temperatures can be adjusted by either increasing the process velocity or by independent change of the nozzle heights. This novel temperature control method shows promise as an adaptive method for rapid control of surface temperatures and was recently patented by the UD-CCM [U.S. Patent No. 5,626,472]. The temperatures are measured and controlled with an AGEMA Thermal Imaging camera with a neural network based PID control system. The camera measures the viewable peak temperatures on the laminate surface and adjusts the nozzle heights accordingly to compensate for any deviation in set point temperatures. The Labview™ interface is also used to input number of layers, ply orientation, surface temperatures, panel geometry, and process velocity. This interface can also be used in conjunction with a laser displacement unit to measure warpage during processing.

Test Materials

The ATP process for thermoplastics lays down prepreg tows, typically 0.125 – 0.2 mm in thickness, with the tow width depending on the hardware. The ATP head at UD-CCM can lay down 6.25-mm wide tows, while industrial machines, such as Cincinnati Milacron's Gantry System can lay down tows as wide as 150 mm.

Fiber reinforced ceramic materials for this study was supplied by ACR in the form of green tows. The reinforcing fiber was carbon and the matrix was zirconium carbide or silicon carbide based, in a low temperature thermoplastic binder. The tows were thicker than the thermoplastic tows for polymer matrix composites.

ATP Process Modification for Ceramic Tow Placement

Several process modifications were carried out to improve the controllability and quality of the rapid prototyping process. . These modifications were implemented to allow for a greater fiber volume loading within the composite matrix and to facilitate extrusion of wide, rectangular green composite tape rather than a filament with a circular cross-section. The rectangular configuration of the green tape is better suited for use in UD's ATP equipment than to the circular filament. A wider tape was chosen to reduce the chances of the ATP head from bending and twisting of the tape during the tape placement, which could lead to defects in the part. Figure 4 shows the ATP head in action during the fabrication of ceramic laminates. Typical microstructures are shown in Figure 5.

The entire robot movement sequence is setup by computer programs developed for the ATP thermoplastic tow placement experiments. The turntable (Figure 6) can be used either horizontally or vertically for flat components or axi-symmetric components respectively. The torch parameters (temperatures, heights, and gas flow rates), consolidation force and head velocity can be controlled on the fly as inputs to the computer program. Final part dimensions and lay-up sequences are also inputs to the program. Once these inputs are given to the program, it operates the robot in automatic mode and lays down the tows as specified. The robot programming software also allows the simulation of the component fabrication steps (Figure 7) that allows the elimination of bad part build strategies prior to the fabrication of the actual component. Green ceramic matrix laminates of any size (within limits of the robotic workcell), fiber orientation and material system can be fabricated by this technique.

During the tow placement process, the roller compresses the composite tow to promote adhesion of the newly placed tow with the previous tow layer. In this process, the controlling factor is the percentage of tow compression rather than the application of pure compression load. To determine a suitable compression percentage rate for the tows, compression tests were performed to evaluate the potential damage to the interface and the fiber tow as a result various compression percentages. For the CMC composite systems evaluated, it was determined that the compression ratios should be between 80-85% of the as received tow thickness.

The upper temperature limit at which the tows should be ATP processed was determined by Thermogravimetric analysis (TGA), shown in Figure 8. If the tows are processed above this critical upper limit, the resultant material weight loss could lead to the degradation of mechanical properties of the tow. For the CMC systems evaluated under this program, ramping tests performed at a rate of 5°C/min up to 500°C determined the 99% weight loss point temperature to be approximately 170°C. Figure 8 also shows weight loss as a function of temperature, up to 500°C.

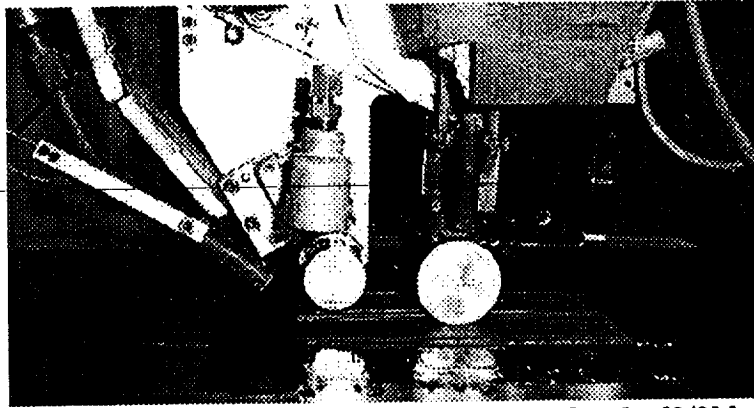


Figure 4. Modified ATP head consolidating 0 ceramic tows for the [0/0] laminate with top 0 layer showing.



Figure 5. Cross-sectional view of the co-extruded matrix-fiber tows coated with a solution-based interface solution. Approximate dimensions of composite tape = 6mm x 0.8mm.

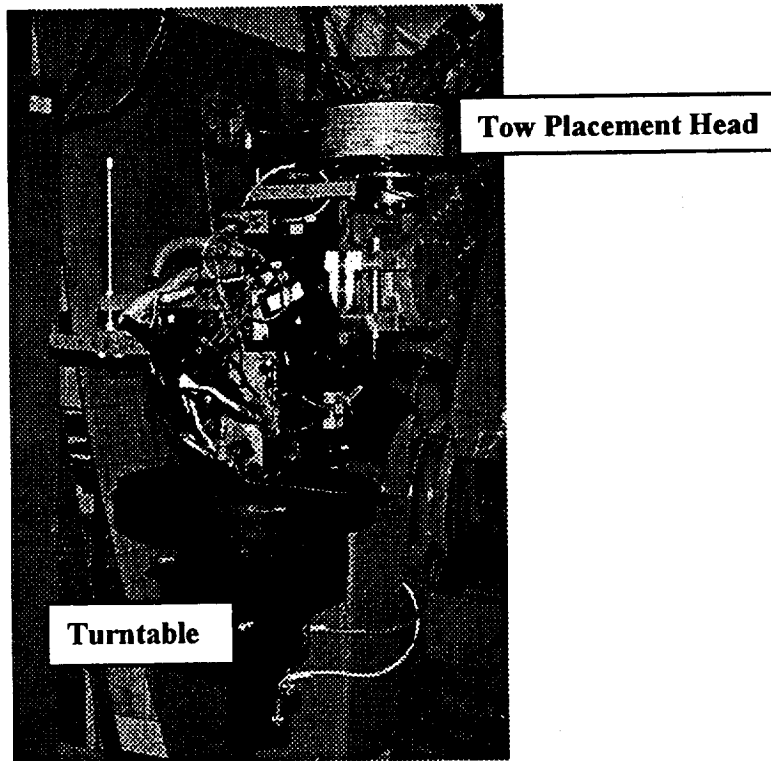


Figure 6. Turntable attached to the ABB robot for fabrication of CMC components

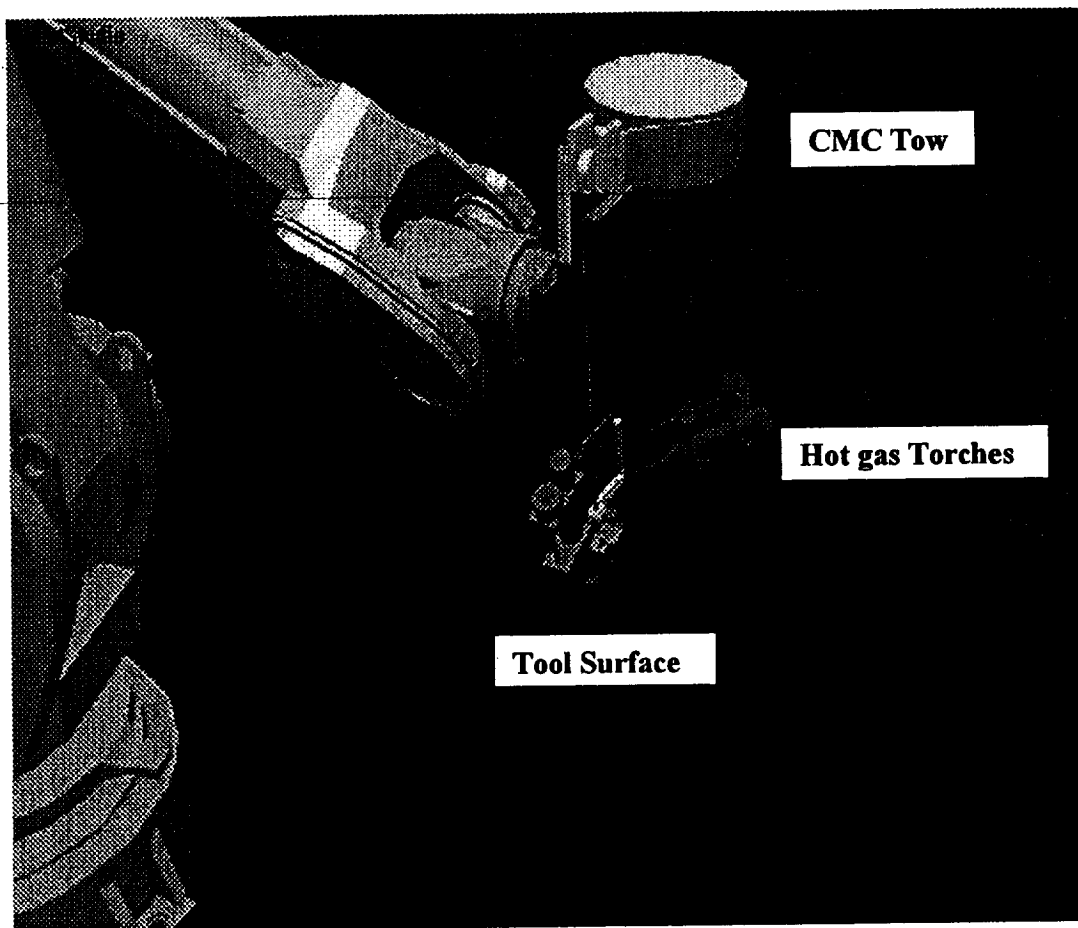


Figure 7. A simulation of the tool path from Robotstudio software.

To determine the weight loss of the tow as a function of time, isothermal TGA tests were performed at 150°C, 160°C, and 170°C. For each temperature, at least 5 samples were tested. The average of all the curves for each temperature is shown below in Figure 9. For 150°C test the final weight was approximately 97.3% of the original material. The final weight values were approximately 97.25% and 97.0% for 160°C and 170°C isothermal tests, respectively.

To determine the lower bound of the heating temperature for ATP processing Differential Scanning Calorimetry (DSC) testing was also performed. The T_g point for the binder systems evaluated under this program was determined to be 93°C. A representative curve from the DSC testing is shown below in Figure 10.

Other components

Figure 11 and 12 show a blisk component being fabricated using the ATP equipment. Figure 13 shows a 6-inch diameter cylinder that was fabricated using the ATP equipment.

Scale-Up Potential

Figure 14 shows the scale-up of thermoplastic ATP process developed in a DARPA RAPTECH (Rapid Technology) program led by UD-CCM with DuPont, Hercules/Alliant and Cincinnati Milacron. In this effort, UD-CCM demonstrated lab-scale viability and developed

optimal process conditions for scale-up. The infrastructure for large parts using the gantry/robotic system exists for large-scale fabrication.

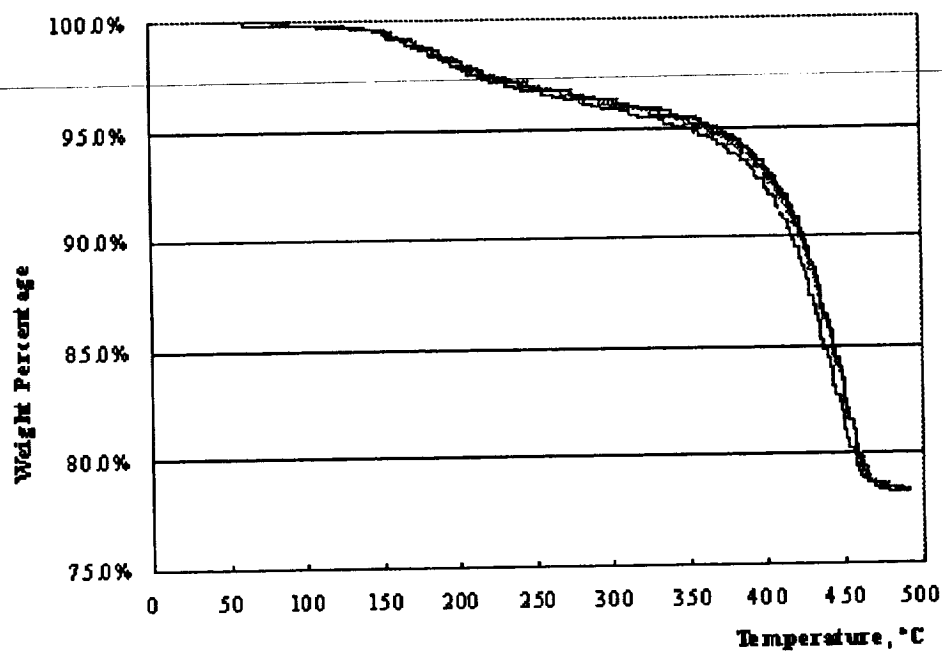


Figure 8. TGA test-weight loss as a function of temperature up to 500°C.

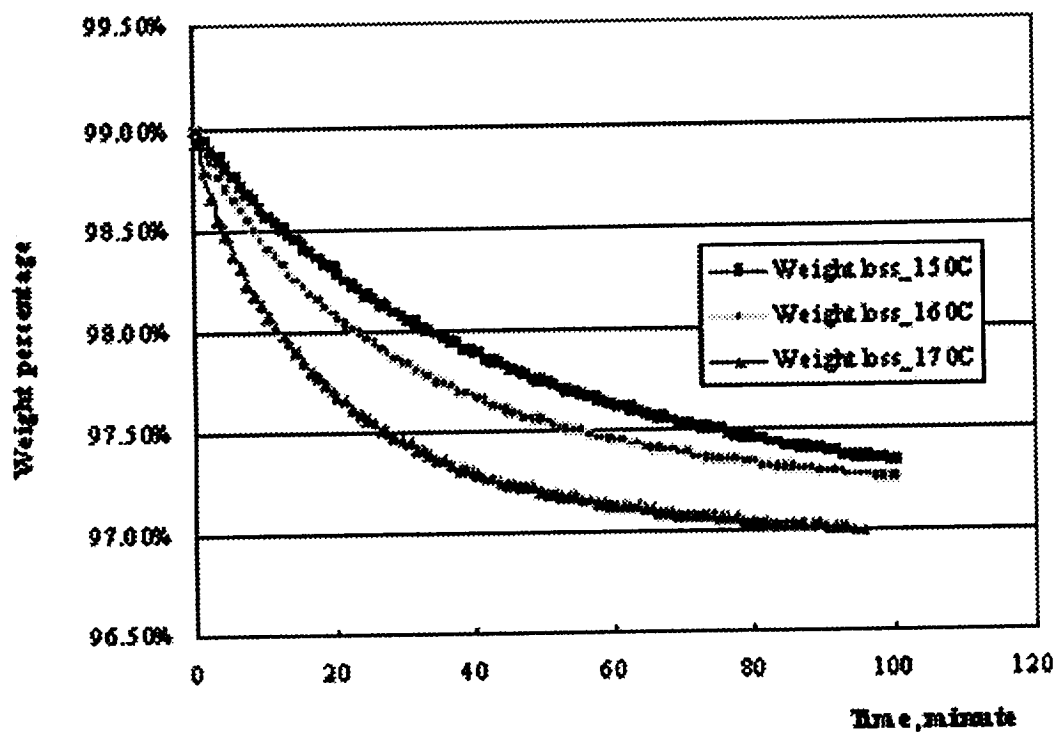


Figure 9. The isothermal TGA tests as a function of time

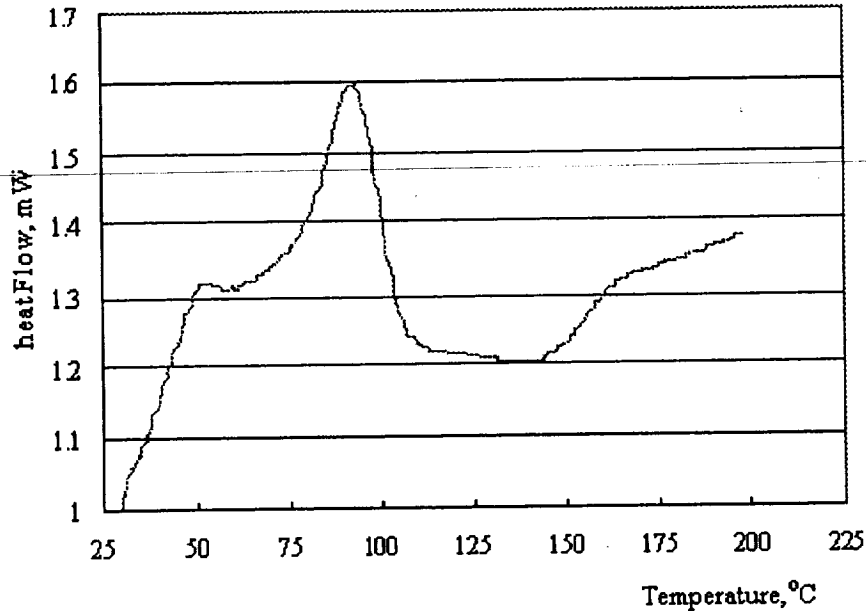


Figure 10. Typical DSC test curve for the ACR CMC tow.



Figure 11. A view of the turntable while fabricating an axi-symmetric component.

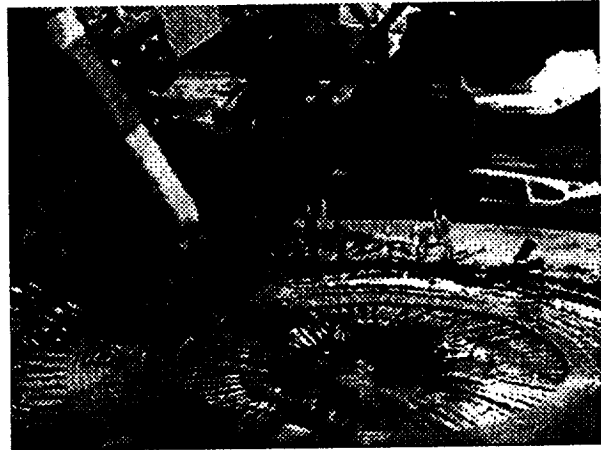


Figure 12. Close up view of the fabrication process for the axi-symmetric component.

Critical Process Parameters

Based on the above results, the critical process parameters were identified to be: (a) consolidation force for each layer, (b) process velocity, (c) torch parameters, and (d) reduction of residual stresses due to the layered manufacturing process. Other important process parameters were identified to be: (a) Redesign of cut and re-feed mechanism, (b) Avoiding the gaps and obtaining proper overlaps, and (c) Refine hardware and electrical set-points to work with CMC type of materials.

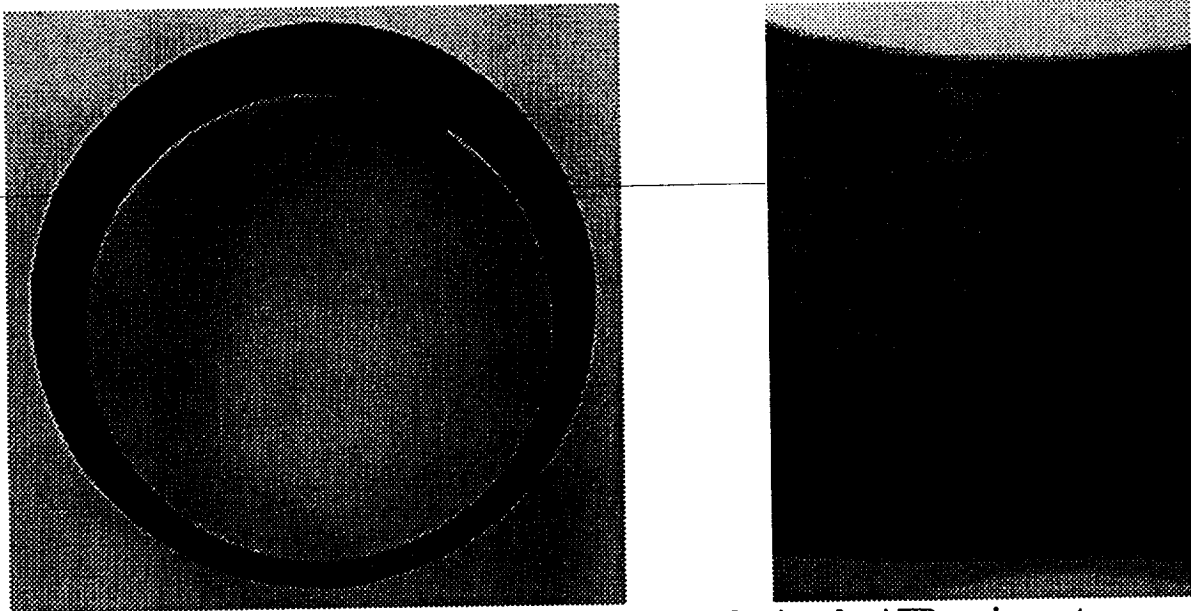


Figure 13. A six inch CMC cylinder fabricated using the ATP equipment

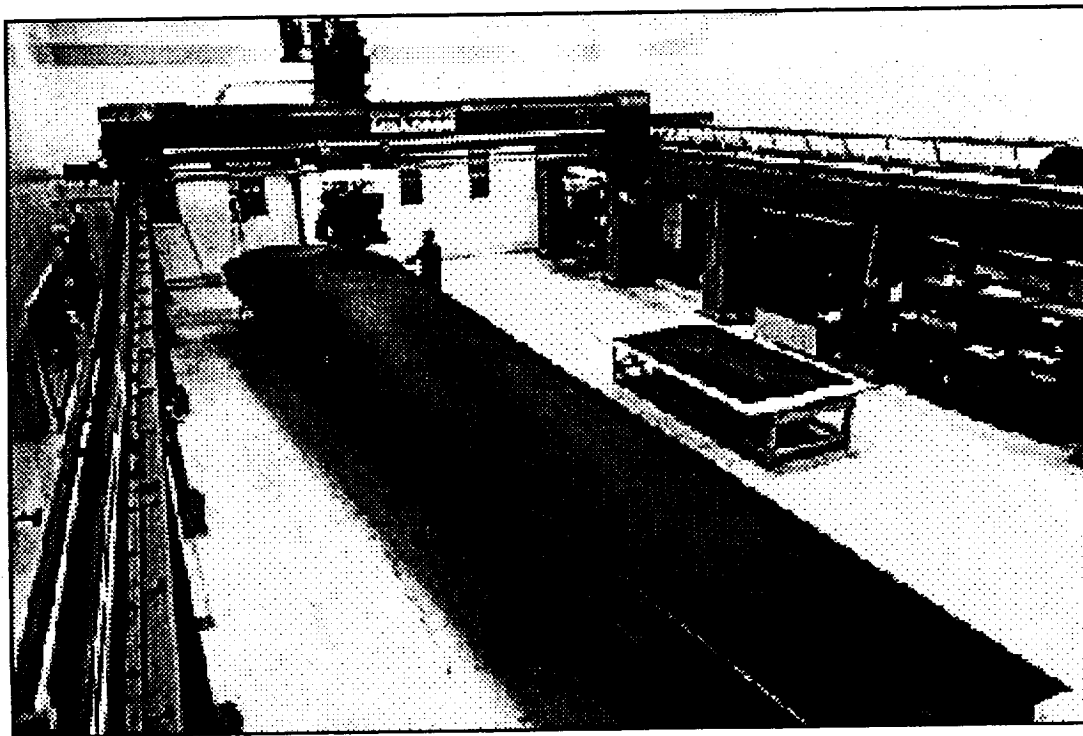


Figure 14. Automated Tape Placement head and gantry. (Picture courtesy of DuPont/Cincinnati-Milacron)

Mechanical property and microstructural characterization of panels

Rapid prototyped CMC billets were subjected to binder burnout and subsequent hot pressing and sintering. Optimization of the interface and the processing conditions are still being carried out. Mechanical test specimens were subjected to 4-point bend testing. All the specimens

exhibited good composite behavior, clear fiber pullout and in some cases, a stable loading pattern, with the load being taken up entirely by the fibers. A typical stress-displacement curve is shown in Figure 15, which shows good load transfer and the ability of the fibers to carry the load after the initial matrix has cracked. In many cases, the composite did not fail entirely and the fibers continued to carry the load. The test had to be discontinued at this point.

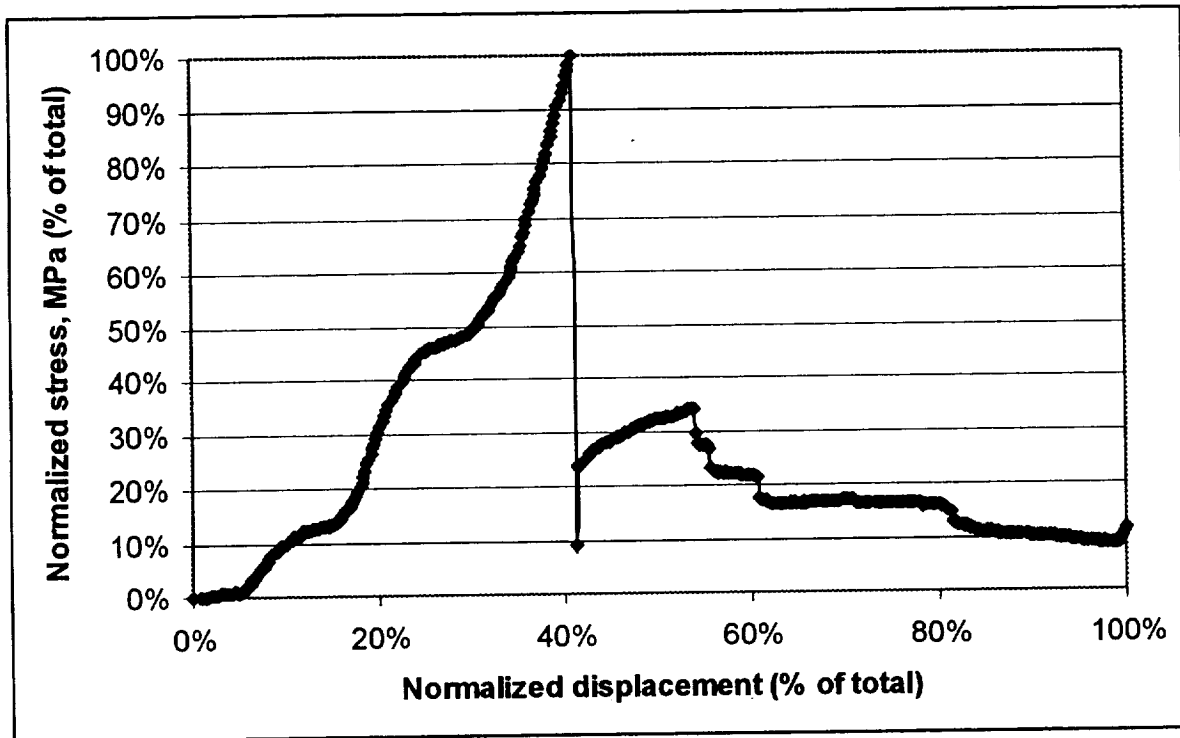


Figure 15. A typical stress-strain curve for a rapid prototyped CMC specimen showing load transfer and the ability of the fibers to carry the load.

LESSONS LEARNED

1. An innovative Ceramic Composite Automated Tow Placement (CCATP) process was successfully developed.
2. Critical process parameters were identified to be: (a) Consolidation force for each layer, (b) process velocity, (c) torch parameters, and (d) reduction of residual stresses due to the layered manufacturing process.
3. Other important process parameters were identified to be: (a) Redesign of cut and re-feed mechanism, (b) Avoiding the gaps and obtaining proper overlaps, and (c) Refine hardware and electrical set-points to work with CMC type of materials.
4. 2D panels were rapidly prototyped using a fiber placement system similar to commercially available fiber placement equipment. Other components were also fabricated.
5. The composite exhibited good fiber pull-out and load-transfer from the matrix to the fibers, although further optimization of the interfacial properties is required to obtain satisfactory mechanical properties.

ACKNOWLEDGMENTS

This work was performed as part of the "Rapid prototyping of continuous fiber reinforced ceramic composites," SBIR program funded by NASA Marshall Space Flight Center under contract numbers NAS8-00084 and NAS8-00192.

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